APPENDIX III

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Flow Analysis

In sheared suspensions at steady state in couette flows the motion of ellipsoids has been mathematically described by Jeffreys[5] and experimentally verified by Mason.[6,7,8] These descriptions give the motion in terms of the angles ϕ and λ shown in Figure 1. If steady state is assumed, then in rotational diffusion the probability density functions for distribution in the angles ϕ and λ are inversely proportional to the angular velocity w of the particles. Hence

$$p(\phi) = \frac{\text{const}}{w\phi}$$

$$p(\lambda) = \frac{\text{const}}{w\lambda}$$

Now given that the particle is orientated somewhere in the field

$$\int_0^{\pi}\int_0^{2\pi} p(\phi)p(\lambda)d\phi d\lambda = 1; \qquad \text{(A1)}$$

the conductivity in a given direction may be then found as

$$\bar{K}_{\alpha} = \int_{o}^{\pi} \int_{o}^{2\pi} p(\phi)p(\lambda)K_{\alpha\alpha}{}^{1}(\phi,\lambda)\,d\phi d\lambda. \quad \text{(no sum)} \qquad \text{(A2)}$$

If the particles are randomly distributed then $p(\phi) = \frac{1}{2\pi}$ and $p(\lambda) = \frac{1}{\pi}$. From geometry the angle θ_3 can be written in terms of ϕ and λ since

$$\cos^2 \theta_3 = \frac{\tan^2\lambda}{\tan^2\phi} . \qquad \text{(A3)}$$

Then

$$\bar{K}_r = \int_{o}^{\pi} \int_{o}^{2\pi} \left[K_b + (K_a - K_b)\cos^2 \theta_3 \right] \frac{1}{2\pi} \frac{1}{\pi}\, d\phi d\lambda \qquad \text{(A4)}$$

$$\bar{K}_r = \frac{K_a + K_b}{2} \qquad \text{(A5)}$$

Given the random uniform dispersion of ellipsoids the dielectric constant measured will be assumed to be $\bar{K}_r$ given by this equation. It should be noted that the dielectric constant of a suspension of axially symmetric ellipsoids which are uniformly dispersed at random orientations is the average of the dielectric constant when aligned. This is independent of whether they are oblate or prolate ellipsoids. This average quantity which is easily measured can be used for determining the shape of particles and for estimating K_a and K_b and hence the relative dimensions of particles.

In a shear flow the particles are not randomly distributed in orientation and do not rotate at a constant angular velocity. The calculation of the dielectric constants then requires a knowledge of the distribution probabilities $p(\phi)$ and $p(\theta)$. These probabilities can be calculated from Jeffreys analysis by using the relation between the angles or orientation and the orbits constant C, for a particle.

$$\tan \theta_2 = \frac{C\, r_e}{(r_e^2 \cos^2\phi + \sin^2\phi)^{1/2}} \qquad \text{(A6)}$$

Where r_e is the axis ratio of the ellipsoid of revolution. The constant (C) of the orbit is determined by the angle at which the axis of the ellipsoid is orientated to the plane of shear and its distribution in a random dispersion would be uniform. In a shear situation there is no theoretical reason, in the limit of Stokes flow, that this will change.

The conductivity for a fixed orbit constant C may then be calculated as follows:

If the effect of Brownian motion on the particle distribution is neglected, then in the steady state

$$p(\phi) = \frac{\text{const}}{w\phi}$$

$$p(\theta_2) = \frac{\text{const}}{w_{\phi 2}}$$

The angular velocity of particles in Couette shear flow (derived theoretically by Jeffreys using Stokes flow approximations) may be expressed for the shear flow shown in Figure 1 as:

$$w_{\theta_2} = \frac{G}{4} \frac{re^2 - 1}{re^2 + 1} \sin 2\theta_2 \sin 2\phi \qquad \text{(A7a)}$$

$$w_{\phi} = \frac{G}{re^2 + 1} \left[re^2 \cos^2 \phi + \sin^2 \phi \right] \qquad \text{(A7b)}$$

for a symmetrical ellipsoid. Using the condition from Equation (A1) together with Equation (A7b) gives

$$p(\phi) = \frac{r_e}{2\pi (r_e^2 \cos^2\phi + \sin^2\phi)} \qquad \text{(A8)}$$

Let us first consider the dielectric constant in the X_2^1 direction (parallel to shear planes, perpendicular to the flow).

From Equation (8b)

$$K_{22}^1 = K_b + (K_a - K_b) \cos^2 \theta_2$$

this can be expressed as a function of the angle ϕ by using Equation (A6) expressed in the form:

$$\cos \theta_2 = \left(1 - \frac{\frac{C^2 re^2}{re^2 - 1}}{\frac{re^2 (1 + C^2)}{re^2 - 1} - \sin^2 \phi} \right)^{1/2} . \qquad \text{(A9)}$$

Then for a fixed C the dielectric constant K_{11}^{1} can be expressed as

$$K_{22}^{1}\Big)_{C} = \int_{0}^{2\pi} p(\phi)\ K_{22}^{1}\ d\phi$$

Integrating using $p(\phi)$ from Equation (38) gives:

$$K_{22}^{1}\Big)_{C} = K_b + (K_a - K_b)\left(\frac{1}{(1 + C^2)[1 + (Cre)^2]}\right)^{1/2}$$

$$= K_b + (K_a - K_b)\ F_2. \qquad \text{(A10)}$$

The dielectric constant for a suspension with the orbits (C) distributed with a probability distribution p(C) can be found as

$$\overline{K_{22}^{1}} = \int_{0}^{\infty} p(C)\ K_{22}^{1}\Big)_{C}\ dC$$

$$= K_b + (K_a - K_b)\ \overline{F_2} \qquad \text{(A11)}$$

where

$$\overline{F_2} \equiv \int_{0}^{\infty} p(C)\ F_2\ dC$$

Similarly the dielectric constant in the X_3^1 direction can be found using the relations that

$$\tan \phi = re \tan \frac{2\pi t}{T} \tag{A12}$$

and

$$\tan \lambda = Cre \sin \frac{2\pi t}{T} \tag{A13}$$

Note: T is the period of revolution of a particle.

By solving for $\tan^2 \lambda$ in terms of $\tan^2 \phi$

$\tan^2 \lambda$ may be shown to be equal to

$$\tan^2\lambda = \frac{(Cre)^2 \tan^2 \phi}{(Cre)^2 + \tan^2 \phi} . \tag{A14}$$

Now from geometry

$$\cos^2 \theta_3 = \frac{\tan^2 \lambda}{\tan^2 \phi} = \frac{C^2 re^2}{(Cre)^2 + \tan^2 \phi} , \tag{A15}$$

then

$$K_{33}^{\ 1} = K_b + (K_a - K_b) \cos^2 \theta_3$$

$$= K_b + (K_a - K_b) \frac{(C^2 re)^2}{(Cre)^2 + \tan^2 \phi} \tag{A16}$$

$$K_{33}^{\ 1}\Big)_C = K_b + (K_a - K_b) \int_0^{2\pi} p(\phi) \frac{C^2 re^2}{(Cre)^2 + \tan^2 \phi} d\phi \tag{A17}$$

$$= K_b + (K_a - K_b) \frac{C}{1 + C} \quad \text{and} \tag{A18}$$

$$\overline{K_{33}^{\ 1}} = K_b + (K_a - K_b) \int_0^{\infty} p(C) \frac{C}{1 + C} dC \equiv K_b + (K_a - K_b)\overline{F_3} \tag{A19}$$

The dielectric constant in the X_1^1 direction is found using the relations

$$K_{11}^{1} = K_b + (K_a - K_b) \cos^2 \theta_1 .$$

From geometry

$$\cos \theta_1 = \cos \theta_2 \tan \lambda$$

Then using Equation (A9) for θ_2 and Equation (A14) for tan λ

$$K_{11}^{1}\Big)_C = K_b + (K_a - K_b) \int_0^{2\pi} \frac{[(re^2-1)\cos^2\phi+1][(Cre)^2\sin^2\phi]p(\phi)\,d\phi}{[(Cre)^2+(re^2-1)\cos^2\phi+1][(Cre)^2-1)\cos^2\phi+1]} \tag{A20}$$

Integrating with $p(\phi)$ from Equation (38) gives:

$$K_{11}^{1}\Big)_C = K_b + (K_a - K_b) \frac{2(Cre)^2 r_e}{[2(Cre)^2+r_e^2+1]} \frac{3}{[(Cre)^2+1][H-D]} \left[\frac{H+1}{(1-H^2)^{1/2}} - \frac{D+1}{(1-D^2)^{1/2}} \right] \tag{A21}$$

$$K_{11}^{1}\Big)_C = K_b + (K_a - K_b) F_1$$

where

$$D \equiv \frac{(Cre)^2 - 1}{(Cre)^2 + 1}, \qquad H \equiv \frac{re^2 - 1}{2(Cre)^2 + re^2 + 1}$$

then

$$\overline{K_{11}^{1}} = \int_0^\infty p(C)\ K_{11}^{1}\Big)_C\ dC\ .$$

Since K_a and K_b are independent of C and $\int_0^\infty p(C) = 1$ this may be written as

$$\overline{K_{11}^{1}} = K_b + (K_a - K_b)\ \overline{F_1}$$

where

$$F_1 \equiv \int_0^\infty p(C)\ F_1\ dC\ . \qquad \text{(A21a)}$$

The evaluation of the distribution p(C) of the orbit constant for a suspension is next required to determine the dielectric constant. The parameter C may range from 0 to ∞. C = 0 corresponds to the symmetric axis of the ellipsoid being perpendicular to the shear planes and C = ∞ corresponds to the symmetry axis of the ellipsoid rotating in the plane of the flow which corresponds to the ellipsoid having the angle $\theta_2 = 90°$.

No theoretical prediction of the distribution of C with flow rate have been made. Two hypotheses have been proposed, however.

1. Jeffreys assumed that the ellipsoids would eventually rotate in such a way as to minimize the energy dissipation. This theory predicts that

   $p(C) = 1$ for $C = 0$

   $p(C) = 0$ for $C > 0$

All the ellipsoids would be alligned with their symmetrical axes perpendicular to the shear planes if a/b < 1 or parallel to the shear planes if a/b > 1. The conductivity with time across shear planes would approach K_a with $\frac{a}{b} < 1$ or K_b with $\frac{a}{b} > 1$ using this assumption.

2. Eisenshitz[9] assumed that P (C) remained the same whether at rest or in motion, so that every orbital constant C was equally probably.

The first assumption gives the results shown in Figure 3 curve (A). Since it has been demonstrated that Jeffrey's hypotheses is not correct, we will assume a uniform distribution of the orbital constant (the Eisenschitz assumption). In this case the probability that a particle axis is between C = 0 and C is

$$P(C) = \frac{2}{\pi} \int_0^{\pi/2} [1 - \cos \theta_2 (\phi)] \, d\phi \qquad (A22)$$

Substituting Equation (A7) into Equation (A22) gives

$$P(C) = 1 - \frac{2}{\pi} \int_0^{\frac{\pi}{2}} \left(\frac{1 - \frac{re^2 - 1}{re^2} \sin^2 \phi}{1 + C^2 - \frac{re^2 - 1}{re^2} \sin^2 \phi} \right)^{1/2} d\phi \, . \qquad (A23)$$

Since the probability density distribution $p(C) \equiv \frac{dP(C)}{dC}$ is needed, differentiation gives

$$P(C) = \frac{2}{\pi} \frac{C}{(1 + C^2)^{3/2}} \int_0^{\frac{\pi}{2}} \frac{\left(1 - \frac{re^2 - 1}{re^2} \sin^2 \phi\right)^{1/2}}{\left(1 - \frac{re^2 - 1}{re^2(1 + C^2)} \sin^2 \phi\right)^{3/2}} d\phi. \tag{A24}$$

This equation is of the form of an elliptic integral if the coefficients of the $\sin^2 \phi$ terms range between 0 and 1. For $re < 1$ this is not true and a change in form must therefore be made. First the numerator is expanded as:

$$P(C) = \frac{2}{\pi} \frac{C}{(1 + C^2)^{3/2}} \int_0^{\frac{\pi}{2}} \frac{\left(1 - \sin^2 \phi + \frac{\sin^2 \phi}{re^2}\right)^{1/2}}{\left(1 - \frac{re^2 - 1}{re^2 (1 + C^2)} \sin^2 \phi\right)^{3/2}} d\phi$$

Changing the variable by letting $\phi = \frac{\pi}{2} - \psi$ gives:

$$P(C) = \frac{2}{\pi} \frac{C\, re^2}{[(Cre)^2 + 1]^{3/2}} \int_0^{\frac{\pi}{2}} \frac{[1 - (1 - re^2) \sin^2 \psi]^{1/2}}{\left(1 - \frac{1 - re^2}{1 + (Cre)^2} \sin^2 \psi\right)^{3/2}} d\psi \tag{A25}$$

This is a complete elliptic integral of the second kind.

$$P(C) = \frac{2}{\pi} \frac{Cre^2}{[(Cre)^2 + 1]^{3/2}} \left[\frac{1}{\left(1 - \frac{1 - re^2}{1 + (Cre)^2}\right)^{1/2}} \right] E\left[\left[\frac{\pi}{2}, C\left(\frac{1 - re^2}{1 + C^2}\right)^{1/2}\right]\right]$$

$$= \frac{2}{\pi} \frac{2\ Cre}{[(Cre)^2 + 1](1 + C^2)^{1/2}} \left[E\left[\frac{\pi}{2}, C\left(\frac{1 - re^2}{1 + C^2}\right)^{1/2}\right]\right] \quad \text{(A26)}$$

The other case of interest, i.e., re > 1 are found directly from Equation (A26) as:

$$P(C) = \frac{2}{\pi} \frac{Cre}{(1 + C^2)[(Cre)^2 + 1]^{1/2}} E\left[\frac{\pi}{2}, \left[\frac{C^2(re^2 - 1)}{(Cre)^2 + 1}\right]^{1/2}\right] \quad \text{(A27)}$$

There are two cases where these integrals can be expressed as simple closed solutions.

1. The first is the case where re = 1. This corresponds to a sphere (Hydrodynamically). For this case

$$P(C) = \frac{C}{(1 + C^2)^{3/2}} \quad \text{(A28)}$$

2. For the second case re = ∞ (an infinitely long rod).

$$P(C) = \frac{2}{\pi(1 + C^2)} \quad \text{(A29)}$$

The factors F_i are then calculated using the probability distributions with Equations (A11), (A19) and (A21a). The results are shown plotted in Figure 3.

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